Particle Emission from Laser Printers

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SUMMARY
Recent studies have shown that the operation of laser printers can result in the emission of high concentrations of ultrafine particles. However, fundamental gaps in knowledge still remain, for example, it is not clear what makes a printer a high emitter or why some models alternate between being low and high emitters. In order to provide insight into the formation mechanisms of these particles, comprehensive investigations into particle emissions from one high and one low emitting printer were undertaken. Each printer was operated using its own toner, and printed on two types of paper. Emissions from the printers were studied in a flow tunnel and a box chamber, while emissions from the fuser rollers, two types of paper and toner were investigated in a furnace. This paper provides examples of preliminary results from the study, taken from the extensive body of data which has been collected and analysed so far.

KEYWORDS
Printer emissions, Ultrafine particle emissions, VOC emissions, Printer emitted particle characteristics

INTRODUCTION
Since the publication of the paper by He et al. (2007), demonstrating that about 30% of commonly used printers emit large numbers of ultrafine particles, and that these emissions can results in significant elevation of particle number concentrations in offices where the printers are operated, issues regarding printer emissions have attracted worldwide attention. Before this publication, there were only a handful of papers indicating the possibility of ultrafine particle emissions from printers (Brown, 1999; Stefaniak et al., 2000; Tuomi et al., 2000; Lee et al., 2001; Wensing et al., 2002; Namiki et al., 2006; Wensing et al., 2006), supported by a few other very recent publications (Kagi et al., 2007; Destaillats et al., 2008). These studies indicated that particles are not the only pollutants emitted by printers, and indicated that printers also emit VOCs and ozone. It has been shown that there are potentially many factors that may affect printer emission rates and other emission characteristics including: printer model, printer age, cartridge type and cartridge age, as well as paper type. However, the relationship between these factors and the emission characteristics of pollutants has not been established. While current research in relation to ultrafine particles (which are considered a particular risk to human health due to, among other things, the potential to penetrate deep into the lung and to translocate to other parts of the body) has demonstrated that the operation of some printers results in emission of high concentrations of ultrafine particles, fundamental gaps in knowledge in relation to the emissions still remain. Firstly, there is only some insight into the size distribution of the particles, while there is little

knowledge of other properties of the particles, including chemical composition. Secondly, the formation mechanisms of the particles resulting from printer operation are not clear. Due to these gaps in scientific knowledge, there is a considerable lack of answers to some very practical questions, such as what makes a printer a high emitter or why some printer models are sometimes low emitters and sometimes high emitters. It is of critical importance to find answers to such questions, as this would enable more targeted protection against exposure to printer emissions, and more importantly, it would enable the design and manufacture of printers that would minimise or eliminate the emission of particles. Therefore the aim of this work was to undertake the most comprehensive investigations possible, in order to develop an understanding of particle formation mechanisms in laser printers, as well their chemical and physical characteristics. The aim of this paper is to discuss the experimental approaches to this research, as well as present some preliminary conclusions.

EXPERIMENTAL METHODS

A number of different instrumental techniques were utilized to fulfil the aim of this work, with many of them requiring special experimental set ups and particular operating conditions. In general, three main types of experiments were conducted: (1) Controlled furnace heating of three key components of the printing process (printer paper, toner powder and fuser rollers), to individually investigate the response of these to elevated temperature; (2) Emission testing of printers located in a flow tunnel, to quantify particle emission factors and to study the emission products under dynamic conditions, with minimum interaction between the products and the enclosed environment; and (3) Emission testing of printers located in a box chamber, to study those elements of the process that require a longer sampling time and therefore can not be studied in the flow tunnel (due to the longer residence time of emission products in the chamber, interactions between the products and the walls of the chambers had to be considered).

What was tested?

It was decided that, based on the results of previous work by the authors He et al. (2007), two common printers would be selected for this work: Printer A being a high emitter and Printer B being a low particle emitting printer. Each printer was operated using its own toner, and printed on two types of paper, each being 80 grams per m^2. Emissions from the operating printers were studied in the flow tunnel and the box chamber, with the printers operated to generate 0%, 5% or 50% toner coverage. Emissions from the fuser rollers of each printer, as well as from the two types of paper and samples of the toner were investigated in the furnace.

Main instrumentation, sampling and analytical methods used

- Particle size distribution and concentration: Two TSI Incorporated (St. Paul, MN) Condensation Particle Counters (CPCs) were used for measurements of particle total number concentration: a TSI Model 3022A and a TSI Model 3781 CPC, with a sampling time of 2 s, and size range for 0.007 - 3 μm and 0.006 - 3 μm, respectively. A TSI Model 8525 P-Trak Ultrafine Particle Counter was used to measure total particle number concentration (sample time 1 second) in the size range 0.02 - 1 μm. Particle size distribution in the submicrometer range was measured by a Scanning Mobility Particle Sizer (SMPS) comprising of a TSI Model 3080 Electrostatic Classifier (EC) and a TSI Model 3025A CPC, with a sampling time of 120 s or 180 s and size range of 4 - 160 nm, and particle size distribution in the supermicrometer range was measured by a TSI Model 3320 and a TSI Model 3312A Aerodynamic Particle Sizer (APS), with a sampling time of 10 s and the size range of 0.54 - 20 μm. Particle mass concentration was measured by a TSI Model 8520 DustTrak Aerosol Monitor using a 2.5 μm impactor at the aerosol inlet.

- Particle volatility and hygroscopicity: A Volatility and Hygroscopic Tandem Differential Mobility Analyser (VH-TDMA) (comprising of two TSI Model 3010 CPCs and three TSI Model 3080 ECs) was used to measure the thermal decomposition and hygroscopic behaviour of particles in the dominant size range. The VH-TDMA selects particles in a well defined size range, 0.007 - 0.7 μm, and measures the volume of each distinct volatile species, along with its water activity (hygroscopic growth) at each stage of the volatilisation process (Johnson et al. 2004).
- Total Volatile Organic Compounds (TVOC): A PPB Rae Plus Photoionisation Detector (PID) was used to monitor real time TVOC. This was done for preliminary assessments of VOC background concentration prior to commencement of experiments.
- Temperature during printing: A thermocouple bead was inserted through the back of the printer and placed in contact with the fuser roller during printing. It had to be withdrawn just prior to the paper completing its passage over the roller to prevent the thermocouple lead from getting entangled.
- Furnace: A standard electric tube furnace with sillimanite linings and a manual temperature control was used. The investigated samples were placed in a ceramic container located in the middle of the furnace. The temperature of the samples was monitored by thermo-coupling.
- Flow tunnel: The time dependency of the emissions was investigated by placing the printer in a flow tunnel, which is essentially a small wind tunnel through which filtered air is propelled by a HEPA-filter/fan module past the printer at a low, controlled velocity. These emissions were carried by the airflow to instruments located downstream. Air velocity in the tunnel was measured by a TSI Velocicheck Model 8330 Anenometer.
- Box chamber: The wooden box chamber used for the study was $1m^3$ in volume, with the inner surface painted with three layers of latex paint.

Samples were collected for the following type of analysis:

- Volatile and Semivolatile Organic Compounds (VOCs and SVOCs): Active sampling was conducted on stainless steel desorption tubes (Perkin Elmer) that were filled with Tenax TA (Chrompack), using Chematec FLEC pump which operated at 150 ml min^{-1}, for 34 min. They were subsequently analyzed in the laboratory by a process including tube thermal desorbtion (320°C, 10 min; Perkin Elmer ATD 400) into a GC/MS system (Agilent 6890/5973). The compounds were separated on a HP-5 MS column (60 m x 0.25 mm, 0.25 μm). Initial qualitative analyses were based on a PBM library search (McLafferty and Turecek 1993), with confirmatory analyses using mass spectra and retention data obtained from authentic compounds.
- Individual particle chemical composition and morphology: Particles were collected on TEM grids, coated with thin films of carbon or Formvar polymer and were investigated by Transmission Electron Microscopy (TEM).
- Analyses of paper and toner: To identify potential particle emission sources from these materials, small samples of printer Paper 1 and the toner powder for Printer A were examined in an FEI Quanta Scanning Electron Microscope (SEM) by secondary and backscattered electron imaging, and small particulate constituents of the samples were analyzed by EDX for element composition.

Study design

- Furnace and box chamber: The two types of printer paper, along with samples of toner powder from the two toner cartridges and two fuser rollers were individually placed in a ceramic container and heated in the furnace. The temperature of the furnace was increased at a rate of approximately 0.5°C min^{-1} up to 150°C, 160°C and 210°C for the paper, rollers and

toner powders, respectively, and maintained at the highest temperature for approx. 20 minutes and then decreased back to normal room temperature. Sampling of the emissions was conducted according to the schematic diagram presented in Figure 1. It can be seen that particle number and size distribution was measured directly from the furnace, while other emissions were tested from the box chamber, to which air was introduced from the furnace at a flow rate of 1.4 L min^{-1} During the cooling of the furnace, nitrogen instead of compressed air, was introduced at a rate of 0.3 L/min, until the temperature reached room temperature (approx. 24°C), in order to minimise VOC adsorption onto the walls of the furnace and thus the contamination of subsequent experiments. Prior to sampling, the furnace was cleared of VOCs by heating at 300°C for 8 hours and the box chamber was cleaned using distilled water. Particle free air was then introduced into the furnace and the box chamber by passing compressed air through a HEPA filter at a flow rate of 2 and 8 L min^{-1}, respectively. These steps were taken to achieve and maintain acceptable low background contaminant concentrations. For the box chamber, acceptable background real time TVOC, particle number and $PM_{2.5}$ concentrations at the commencement of the experiment were 5 ppb, < 10 particles per cm^3 and < 0.001 mg m^{-3}, respectively.

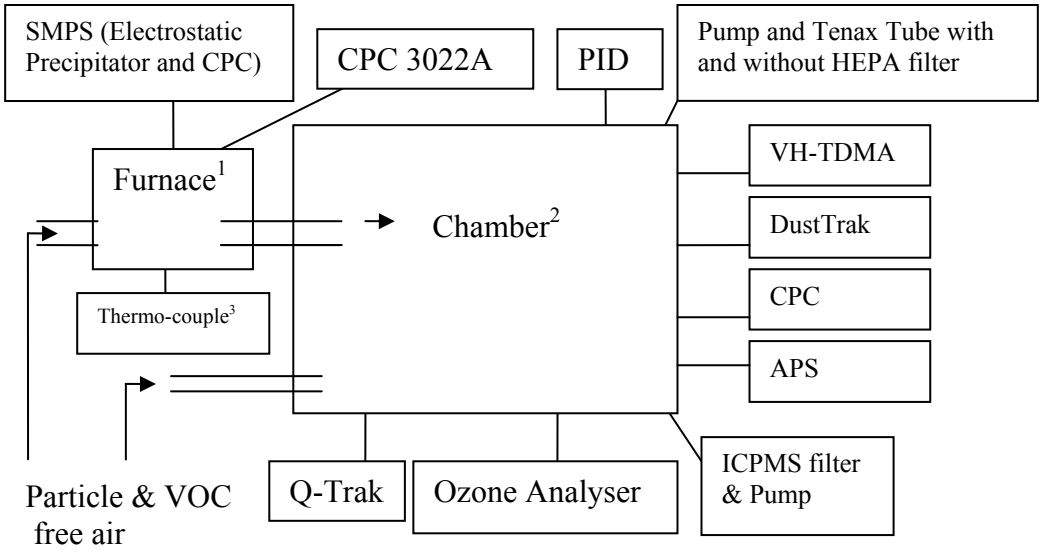

Figure 1. Schematic diagram of the instrumental set up for furnace and box chamber measurements. [1] Toner, fuser roller or paper in-situ, [2] Printer in-situ (without furnace), [3] Thermo-couple to object

- Box Chamber: The box chamber was used to collect emissions from printers in operation. The printer was placed in the box chamber without the furnace connected to the chamber. During each test 150 pages were printed. Air from the chamber was sampled and analyzed as per Figure 1.
- Flow tunnel: The flow tunnel was also used to collect emissions from printers in operation. Air velocity in the tunnel was 0.7 m s^{-1}. During each test 150 pages were printed and after each printing episode, the printers were allowed between 30 and 60 minutes to cool down before repeating the experiment for different type of paper and/or level of toner coverage.

RESULTS

The experimental results consist of a very large body of data for all three types of experiments (furnace, flow tunnel and box chamber). It is outside the scope of this paper to provide a complete set of the experimental data or in depth analysis of the entire set. Instead, examples

of some results are provided, followed by a discussion of some elements of the particle emission process.

Particle size distribution measured during heating of the toner powder in the furnace

Figures 2 and 3 present particle size distribution measured during heating of toner powder A and B, respectively. It can be seen that both particle size distributions are lognormal and of a very similar CMD (10.2 nm and 12.2 nm, respectively). The volatilization and humidification experiments also showed that the volatilisation temperature remained similar for particles of comparable size (Figure 3). However whilst these particle were found to be volatile, they showed no growth when exposed to high humidity and were therefore non-hygroscopic.

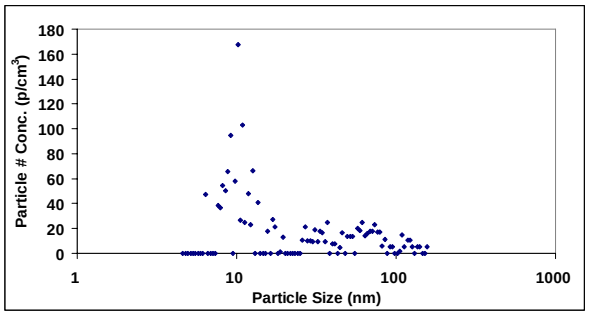

Figure 2. Particle size distribution resulting from heating of toner powder A in the furnace.

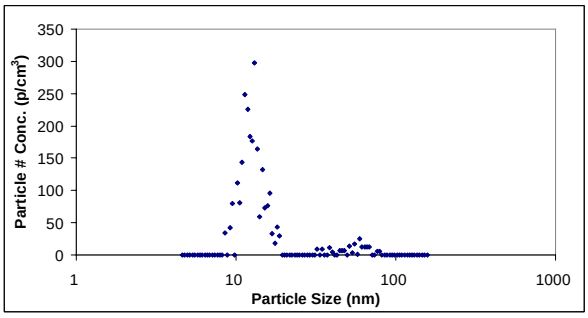

Figure 3. Particle size distribution resulting from heating of toner powder B in the furnace.

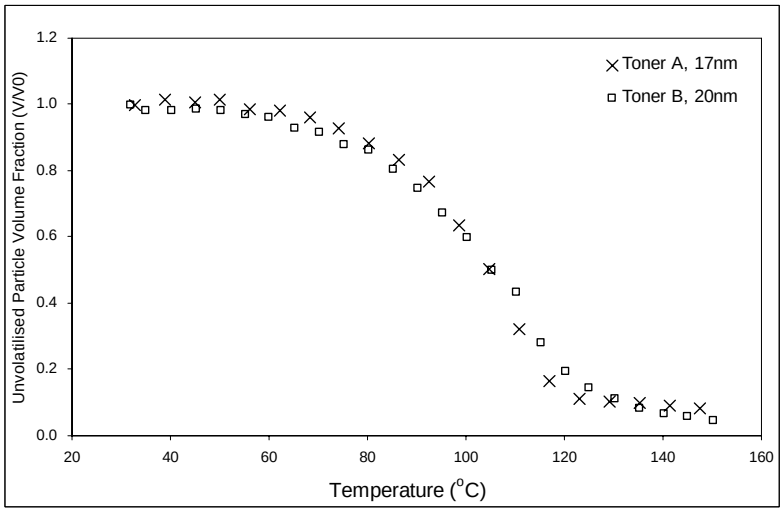

Figure 4. Volatilisation temperature curve of the toner generated particles, measured by the VH-TDMA (initial particle sizes are given in the legend).

Emissions from printers operating in the flow tunnel

Printers A and B were operated in the flow tunnel using Paper 1, at three different levels of toner coverage. Some of the results for Printer A (high emitter) are presented in Figures 4 and 5. Figures 5 and 6 present the submicrometer (CPC) and supermicrometer (APS) particle number concentration, and TVOC and ozone, during and after the conclusion of the printing process for 5% print coverage, for Printer A and B, respectively. In each case 150 pages were printed, which took about 7 minutes. Figure 7 presents particle size distribution measured

immediately after commencement of the printing, about 2 minutes later and about four minutes later, respectively.

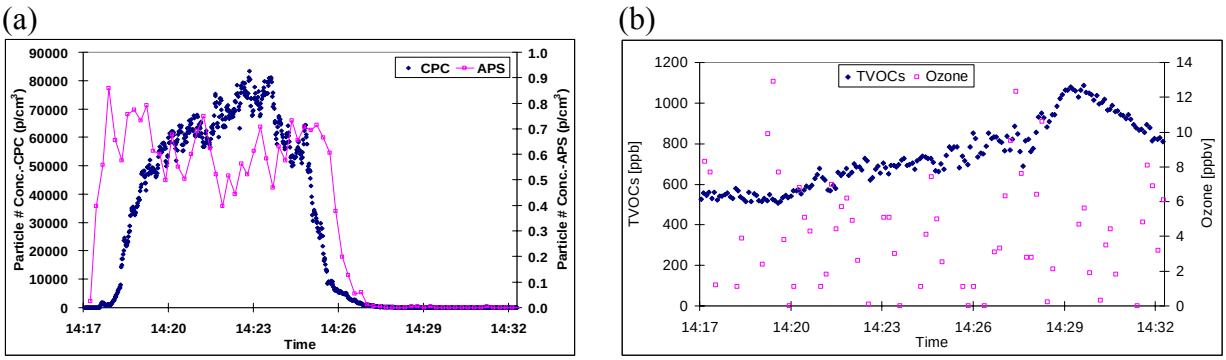

Figure 5. Characteristics of emissions in the flow tunnel from Printer A during and after printing of 150 pages on Paper 1 at 5% toner. (a) Particle number concentration measured by CPC and APS; and (b) TVOCs and ozone concentration.

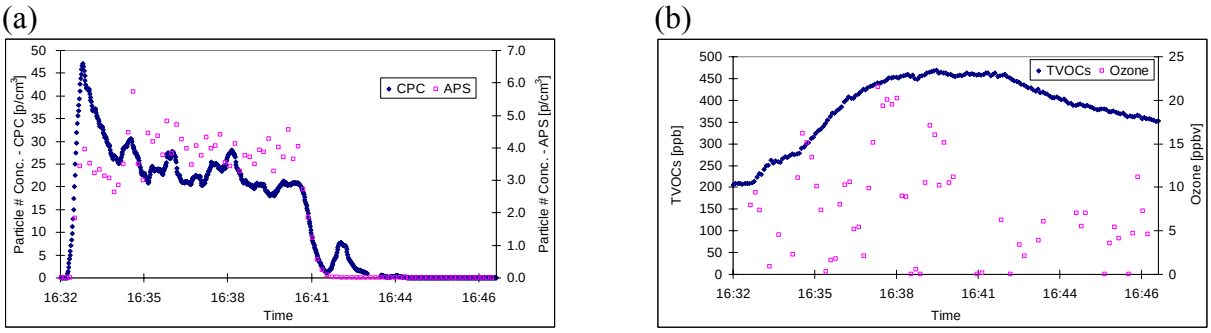

Figure 6. Characteristics of emissions in the flow tunnel from Printer B during and after printing of 150 pages on Paper 1 at 5% toner. (a) Particle number concentration measured by CPC and APS; and (b) TVOCs and ozone concentration.

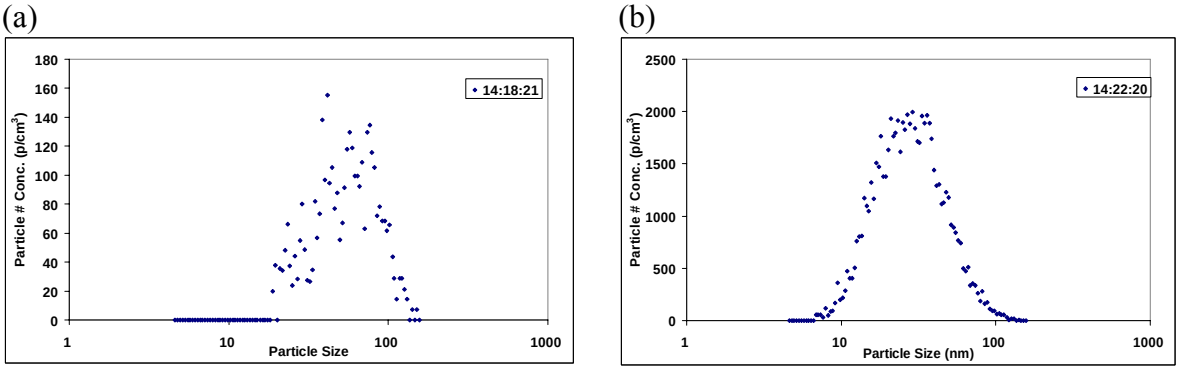

Figure 7. Particle size distribution for Printer A when printing 150 pages on Paper 1 at 5% toner coverage. (a) During the first 2 minutes of printing; and (b) During the last 2 minutes of printing.

DISCUSSION AND CONCLUSIONS

While the large body of data from these most comprehensive experiments aimed at characterisation of particles emitted by laser printers as well as uncovering the mechanisms

responsible for particle generation is still being analysis and different aspects of data pieced together to generate a complete picture, several important conclusions can be derived based of the examples of the data presented above.

Furnace experiments

While only one example of particle characteristics emitted from toner powder (two types) heated in the furnace to a temperature of over 200°C was provided above, this example provides a detailed characterisation of particle, VOCs and ozone emanation from each of the key components of the printing process, including paper, toner powder and fuser roller. In this case, it can be seen that the count median diameter (CMD) of particles resulting from heating of the toner was very similar for both toner powders, of 10.2 nm and 12.2 nm, respectively.

Flow tunnel experiments

- Temperature and RH: For Printer A, immediately after the commencement of the printing, RH in the tunnel start sharply increasing, and keeps increasing until reaching a maximum about 160 seconds later. The overall increase is about 5% RH. Following this, it starts decreasing (despite printing continuing), with the decrease continuing for 290 seconds after termination of printing, when the RH reaches the background level (all the printed pages remain in the tunnel during that time). The temperature in the tunnel also starts increasing shortly after commencement of printing, however, about 50 seconds later than the increase of RH. The temperature increases by 0.8°C, after which it plateaus and starts decreasing about 320 seconds after cessation of printing.
- Particle number concentration: For Printer A, submicrometer particle number concentration starts increasing about 28 s after commencement of printing and continuing until the point of printing cessation. The time series pattern displays first a small, but distinctive peak, followed by a sharp increase in concentration. The highest concentration reached in the tunnel was about $8x10^4$ particles cm^{-3}. Supermicrometer particle concentration displayed a somewhat different trend, with sharp increase in concentration immediately after commencement of printing (sharper and earlier than for submicrometer particles), followed by a rapid decrease in concentration about 40 seconds later, and then by another increase, not always to as high values as before.
- Ozone: Time series for ozone concentration for Printer A does not show any trend in the concentration, with the concentrations remaining at the background values during the entire time of printing.
- TVOCs: For both printers TVOC concentration either remained steady or increased slowly until the end of the printing process.
- Particle size distribution: There was a similar pattern for size distribution changes during printing for all three levels of toner coverage. The first displayed the presence of large particles with the CMD in the range between 60 – 80 nm. The spectra appear to display several peaks, however particle concentration is too low to attempt a modal analysis of the spectra. The measurements of the size distribution conducted 2 minutes later show a bimodal distribution, with significantly smaller particles than in the first spectrum. The final spectrum, 2 minutes later appear to be unimodal.

Box Chamber experiments

These experiments provided, among other things, very important information about particle volatility, pointing out to possible volatile or semi-volatile nature of the particles. It should be kept in mind that the minimum detectable particle size was about 7 nm and therefore, in principle, it cannot be claimed that no unevaporated core remained.

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